U.S. DEPARTMENT OF ENERGY
OAK RIDGE OPERATIONS
OAK RIDGE ORNL SITE OFFICE

TRANSMITTAL OF INFORMATION

The enclosed is to fulfill a request made by Chem Risk, as part of the *Oak Ridge Health Studies agreement* efforts. These documents have received the necessary reviews and may be released to the Chem Risk.

TIO Release Approval: David R Hamrin

Information enclosed: ORNL/CF-77/58

Requested by: Young Moon

Requested from: Linda Hill

Approved: Timothy W. Joseph
Timothy W. Joseph
Program Manager
DOE ORNL Site Office

Date: April 28, 1997

cc w/o enc: J. L. Weaver, 4500S, MS. 6141.

[redacted]

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OAK RIDGE NATIONAL LABORATORY

OPERATED BY

UNION CARBIDE CORPORATION

NUCLEAR DIVISION

POST OFFICE BOX X

OAK RIDGE, TENNESSEE 37830

77821

OFFICE OF THE DIRECTOR

February 22, 1977

U. S. Energy Research and Development
Administration
Oak Ridge Operations
Attention: Mr. J. A. Lenhard
Research and Technical Support Division
Post Office Box E
Oak Ridge, Tennessee 37830

Gentlemen:

Historical Data

The attached data is furnished in response to your letter dated February 2, 1977, reference OAN:EDM.

These data were developed from ORNL accountability reports submitted for the listed years and are the ORNL official records of transactions.

If you have any questions, please contact H. C. Austin, extension 3-1121.

Very truly yours,

Herman Postma

Herman Postma
Director

HP:HCA:slm

Attachment

Distribution:
1-6. J. A. Lenhard
7. R. F. Hibbs, UCC, Y-12
8. H. C. Austin, FP 17
9. F R. Bruce, FP 36
10. J. H. Gillette, FP 3
11. Herman Postma, FP 37
12. Laboratory Records - RC

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ORNL
CENTRAL FILES NUMBER
ORNL/CF-77/58

DATE: February 22, 1977

SUBJECT: Oak Ridge National Laboratory
Historical Summary of Material Unaccounted For in U-233 and Pu-238 Material Balances (U)

TO: J. A. Lenhard, ERDA, ORO

FROM: Herman Postma 20 20

Distribution

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Classified by: P. S. Baker
Title: Classification Officer
Date: February 22, 1977

Exempt from General Declassification Schedule of Executive Order 11652 - Exemption category 5(B)(3), Topic 146 of OR-CG-2, automatically declassified on (Indefinite).

Classification changed to: Unclassified (level and category)
By authority of: ORO Memo 8-23-94 (classification guide)
[redacted] 9-20-94 ADC or ADD signature (first reviewer) Date
Ted Davis 9-27-94 ADD signature (final reviewer) Date

NATIONAL SECURITY INFORMATION
[redacted] Disclosure Subject to Criminal Sanctions.

[redacted]

[redacted]

OAK RIDGE NATIONAL LABORATORY

HISTORICAL SUMMARY OF MATERIAL UNACCOUNTED FOR IN U-233 AND PU-238 MATERIAL BALANCES

The following table presents the ORNL BPID data for uranium-233 and plutonium-238.

The ORNL facility missions and major operations involving ^{233}U were included in our report number ORNL/CF-76/437 dated December 1, 1976.

Except for the programs in FY 1969 and FY 1972 which contributed to material balance deficiencies, ORNL's primary involvement with ^{238}Pu has been maintaining the "Sales Pool" inventory for sales to licensees and others as approved by ERDA.

Fiscal Year	< > indicates gain (+ to inventory) Uranium-233 (grams ^{233}U)	Plutonium-238 (grams ^{238}Pu to tenths)
1952	2	
1953	< 3>	
1954	1	

Variances are due to rounding.

1955	<93>	

Gain represents the difference between quantities estimated to be contained in HW irradiated thorium based on calculated factors and the quantity recovered during Thorex Process runs to recover approximately 3.5 kg of ^{233}U.

1956	74	
1957	<64>	

During these years, approximately 35 kg of ^{233}U were recovered by the Thorex Process. The variances are attributable to measurement uncertainties and to differences between calculated feed and measured product from the recovery process.

1958	805	

The variance is attributable to:

1) 29% represents the difference between theoretical weights, as calculated by shipper, of material contained in short cooled highly irradiated thorium from Savannah River and the measured input to the Thorex Process.

[redacted]

2) 67% represents the difference obtained on remeasurement of ~33 kg of Thorex end product material utilizing the potentiometric method of analysis, a much more accurate method than the alpha count method previously used.

3) The remaining difference is attributable to measurement uncertainties, unmeasured discards and minor spills.

1959 1,364

Represents a variance experienced in the recovery of ^{233}U from raffinates collected during the short decay thorium processing program and is the difference between the calculated quantity of ^{233}Pa put into decay tanks and the quantity of ^{233}U recovered. The ^{233}Pa was carried on ORNL records as ^{233}U on the assumption it would ultimately decay to ^{233}U. Based on the ^{233}U recovered the ^{233}Pa was apparently overstated which is attributed to uncertainties in sampling and analysis. Due to high radiation levels (10 ci/ml) of the solution, less accurate, remote analytical techniques were necessary. Large dilution factors (up to 10^6) magnified small errors in subsequent steps of the analytical procedure. The solutions could not be mixed long enough to insure homogeneous solution. The low solubility of Pa in acid-deficient media resulted in precipitated Pa-compounds and caused large errors in sampling and analyses.

1960 10

Represents difference between book records and actual measurement of the materials.

1961 60 0.1

The ^{233}U difference is attributable to processing, handling and analytical limits of uncertainties experienced in removing fission products and in concentrating solutions.

The ^{238}Pu difference is due to rounding.

1962 433

During November, 1961, a complete physical inventory was taken of the Thorex Pilot Plant material balance. All ^{233}U greater than 95% enrichment was accumulated in a new calibrated storage tank where it was sparged and measured. This inventory was made by more precise measurement methods than had been feasible in the past. The last completely measured physical inventory was made in November, 1959 and materials more precisely measured have been added to (1.8 kg) and removed from (29.1 kg) the storage facility. The

difference is attributable to measurement uncertainties.

1963 93 -0-

The difference is attributable to uncertainties in weighing and measuring and to handling losses in preparing thoria-$^{233}UO_2$ powder for the Kilorod program utilizing the Sol-Gel Process.

1964 894 -0-

Difference is attributable to analytical differences, weighing and liquid level uncertainties, and differences between input and measured output in purifying approximately 50 kg of ^{233}U (to remove ^{232}U daughters) for use in the Kilorod and other AEC programs.

1965 558 -0-

Weighing, measuring, and analytical uncertainties experienced during the processing of ~25 kg of material for storage at the ^{233}U storage and dispensing facility.

1966 <283> -0-

The gain represents the difference in quantities based on book records and quantities based on chemical analyses. The difference is attributable to sampling methods and variances in volume measurements and analytical techniques.

1967 445 <1.1>

Difference in quantities based on book records and quantities based on chemical analyses. The difference is attributable to sampling methods, variances in volume measurements, and analytical techniques.

The ^{238}Pu difference is due to rounding. During an accounting period, several shipments of less than a reportable unit are made which results in rounding differences at inventory time.

1968 <100> 1.7

Difference in quantities based on book records and quantities based on chemical analyses. The difference is attributable to sampling methods, variances in volume measurements, and analytical techniques.

During the three year period (FY 1966 - 1968) receipts totaled 180 kg; shipments totaled 190 kg; 46 kg were purified by solvent extraction and 40 kg were utilized in the preparation of MSRE salt.

^{238}Pu difference is due to rounding and to handling losses in packing small samples for sale.

1969 512 62.8

The ^{233}U difference is attributable to:

1) Measurement variances and unmeasured discards in clean-up residues during decontamination and subsequent conversion and processing of ~16 kg to 25% UO_2-75% ThO_2 microspheres (397 grams).

2) During fabrication of metal foils, material ignited and burned. The variance represents the difference between measured input and material recovered and is due to material released to off-gas system and adhering to clean-up residues (23 grams).

3) A 92 gram difference is attributed to analytical uncertainties, handling losses and measurement variances in preparing MSRE salts.

The ^{238}Pu difference was experienced in an experimental production of ^{238}Pu microspheres for space application using the sol-gel process flow sheet. The difference between input and measured output is attributable to handling losses, process losses, measurement variances, and dilute wash wastes discharged to the Tank Farm from the process waste catch tanks.

1970 455 2.1

^{233}U variances are attributable to:

1) Limits of uncertainty of isotopic assays in remeasuring ~80 kg of ^{233}U account for 285 grams.

2) A difference of 58 grams was experienced during first and second pass calutron runs for the separation of high purity isotopes. The deficiency is attributable to hold-up and embedment of material in calutron liners and source holders and to measurement variances.

3) The remainder of the deficit is attributable to minor spills, material adhering to clean-up residues and measurement uncertainties in processing, heat treating and carbon coating ~50 kg of ThO/UO_2 microspheres.

1.3 g of the ^{238}Pu deficit represents the difference between quantities contained in scrap and the quantity recovered. The balance of the difference is due to rounding and to routine handling and packing differences.

1971	25	-0-

Represents a difference experienced during first and second pass calutron runs for the separation of high purity isotopes. The deficiency is attributable to hold-up and embedment of material in calutron liners and source holders and to measurement variances.

1972	20	25.3

The ^{233}U difference is attributable to weighing, measuring and analytic limits of uncertainty on materials used in LWBR Development experiments.

The ^{238}Pu deficit represents processing differences in preparing PuO_2 pellets, PuO_2 Mo-coated pellets, and PuO_2 Cm-spiked pellets for use in an Isotopic Fuels Research Program.

1973	57	2.0
1974	43	1.8

^{233}U difference resulted from processing $UO_2(NO_3)_2$ for use in the LWBR-support program and is attributable to measurement uncertainties.

The ^{238}Pu difference represents packing and handling losses and rounding differences.

1975	4	10.1

^{233}U difference is due to rounding.

^{238}Pu represents difference between values based on book records and the results of a physical inventory.

1976	<2>	0.1

Rounding difference.